Title
THE EFFECT OF CRYSTALLOGRAPHIC TEXTURE ON THE MECHANICAL ANISOTROPY OF ZIRCALOY-4 SHEET

Permalink
https://escholarship.org/uc/item/8c34c9pk

Author
Morris, J.W.

Publication Date
1987-08-01
Submitted to Metallurgical Transactions A

The Effect of Crystallographic Texture on the Mechanical Anisotropy of Zircaloy-4 Sheet

S.K. Hwang, J. Lee, and J.W. Morris, Jr.

August 1987

TWO-WEEK LOAN COPY
This is a Library Circulating Copy which may be borrowed for two weeks.

Materials and Chemical Sciences Division
Lawrence Berkeley Laboratory • University of California
ONE CYCLOTRON ROAD, BERKELEY, CA 94720 • (415) 486-4755

Prepared for the U.S. Department of Energy under Contract DE-AC03-76SF00098
DISCLAIMER

This document was prepared as an account of work sponsored by the United States Government. While this document is believed to contain correct information, neither the United States Government nor any agency thereof, nor the Regents of the University of California, nor any of their employees, makes any warranty, express or implied, or assumes any legal responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by its trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof, or the Regents of the University of California. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof or the Regents of the University of California.
The Effect of Crystallographic Texture on the Mechanical Anisotropy of Zircaloy-4 Sheet

S.K. Hwang*, J. Lee* and J.W. Morris, Jr.+ 

* Inha University, Department of Metallurgical Engineering, Incheon, KOREA

+ University of California, Berkeley, Department of Materials Science and Mineral Engineering and Center for Advanced Materials Materials and Chemical Sciences Division Lawrence Berkeley Laboratory Berkeley, California 94720

This work was performed under the auspices of the Korean Science and Engineering Foundation, and partly supported by the Director, Office of Energy Research, Office of Basic Energy Science, Materials Sciences Division of the U.S. Department of Energy under contract # DE-AC03-76SF00098.
The Effect of Crystallographic Texture on the Mechanical Anisotropy of Zircaloy-4 Sheet

S.K. Hwang*, J. Lee* and J.W. Morris, Jr.+ 

ABSTRACT

A quantitative relationship between the crystallographic texture and the mechanical anisotropy of Zircaloy-4 sheet was studied by varying cold working and heat treatment conditions. The relative intensity of the basal poles along the sheet normal direction increased with the amount of cold rolling and increased further by additional α-phase heat treatment. Cross rolling was found to be more effective than direct rolling in enhancing the normal basal texture for the same amount of cold work. β-quenching heat treatment decreased the anisotropy considerably and randomized the pole distribution. The mechanical anisotropy parameter R was obtained from the yield loci constructed by the Knoop microhardness measurements. The R values monotonically increased with the texture parameters fn or Fn obtained by X-ray diffraction method. The qualitative relationship between R and fn or R and Fn followed the trend predicted by a model of an earlier work. Quantitatively, however, the R values showed a positive variance from the theoretical prediction especially for the near-isotropic materials.

* Inha University, Department of Metallurgical Engineering, Inchon, KOREA
1. INTRODUCTION

Nuclear fuel cladding made of Zircaloy may become susceptible to a brittle failure during power ramp or transient situations. The cause of the failure has been attributed to stress corrosion cracking (SCC); the required conditions include the hoop stress due to PCI (Pellet-Clad-Interaction) and an aggressive environment involving iodine in the form of free iodine or ZrI4 gas\(^\text{(1-9)}\). In addition to the mechanical and chemical environment, anisotropy is an important materials characteristic that may alter the iodine-induced SCC susceptibility. There have been observations\(^\text{(10-12)}\) that tubing with higher strength along the radial direction showed a higher resistance to SCC.

The anisotropy of tubing can be measured by a destructive testing, for example, by uniaxial tensile tests. Alternatively, nondestructive methods such as X-ray diffraction can be used to estimate the crystallographic texture which leads to mechanical anisotropy. The latter approach may be more applicable to the advanced method of tubing inspection where individual tubes are nondestructively inspected continuously. Van Swam et al.\(^\text{(13)}\) made an effort to correlate the two quantities in fuel cladding tubing. The objectives of the present work were, first, to determine the conditions of cold working and heat treatment that affect the sheet texture of Zircaloy-4 significantly, and, second, to determine the quantitative relationship between the texture numbers and the anisotropy parameter.

2. EXPERIMENTAL PROCEDURE

The Zircaloy-4 sheet used for the present study was 2 mm thick and recrystallization annealed. The nominal composition range of the sheet was 1.2-1.7 % Sn, 0.18-0.24 %
Fe, 0.07-0.13% Cr, 0.28-0.39% (Fe + Cr) and balance Zr. The yield strength, ultimate tensile strength and elongation of the alloy in the as-received condition were 303 Mpa, 393 Mpa and 15%, respectively. The microstructure in the as-received condition consisted of equiaxed alpha grains approximately 10 μm large. The reference directions of the sheet are indicated in Fig. 1.

Prior to the texture measurement the Zircaloy-4 sheet underwent either cold rolling only or cold rolling and heat treatment. Cold rolling was done by two different methods, direct rolling and cross rolling, at room temperature. In the direct rolling strips were rolled along the original rolling direction (r) the sheet from 2 mm to desired thickness in nine passes, while in the cross rolling the strips were first rolled along the original rolling direction of the sheet to 1.5 mm in four passes, then were turned 90 degree and rolled further to desired thickness in seven steps along the transverse direction (t). The amount of cold work after the final rolling pass was 42.5% and 50%, respectively. All the heat treatments were conducted in approximately 10⁻³ torr vacuum.

The crystallographic texture was analyzed in an X-ray diffractometer with a Cu target. The procedures to obtain the texture parameters, fₙ (Kearns number) and Fₙ (Kallstrom number), are described elsewhere(14-15). The Kearns number and the Kallstrom number were obtained from the inverse pole figure and the direct pole figure, respectively and the numbers represent the relative intensity of the basal poles aligned to the sheet normal. For the inverse pole figure the polished sheet surface was exposed to the X-ray beam so that the specimen normal was identical to the sheet normal. The angular distribution of the poles with respect to the basal poles in the inverse pole figure is illustrated in Fig. 2. Specimens for the direct pole figures were prepared such that the surface normal was inclined to the sheet normal by 54.5 degree. Details of the specimen preparation and X-
ray diffraction technique are described elsewhere\(^{(16)}\). The random texture used for normalizing the diffraction peaks was obtained from a specimen made of filings.

The mechanical anisotropy of specimens was characterized by the anisotropy parameter R. The parameter was obtained by the Knoop hardness method developed by Wheeler and Ireland\(^{(17)}\), which essentially consists of measuring the Knoop hardness on three orthogonal sections making six indentations (two perpendicular indentations on each surface) and calculating R values from the following equation proposed by Backofen et al.\(^{(18)}\):

\[
\sigma_x^2 [1 + \alpha^2 - \alpha (2R/(R+1)) ] = \chi^2
\]  

(1)

where \(\sigma_x\) is the stress along the rolling direction, \(\alpha\) is the ratio of \(\sigma_y\) to \(\sigma_x\) and \(\chi\) is the uniaxial yield stress along the rolling direction. In the actual process the yield loci were obtained from the predetermined \(\alpha\) values and \(\sigma_x\) values computed from the Knoop hardness number and the R values were calculated by using Eq. (1).

3. RESULTS

The texture characteristics of Zircaloy-4 sheet studied were sensitive to plastic deformation and heat treatment. The variation of the pole distribution due to different processing condition was readily discernible from the changes in the relative intensity of poles with respect to a reference orientation, the sheet normal. The three selected inverse pole figures shown in Fig. 3 correspond to the as-received condition, an \(\alpha\)-phase heat treated condition (\(550^\circ\text{C}/1.5\text{ h}\)) after 50 % cold rolling and a \(\beta\)-phase heat treated condition (as-received + \(1000^\circ\text{C}/1.5\text{ h}\)), respectively. While cold rolling and \(\alpha\)-phase heat treatment enhanced the basal texture along the sheet normal direction, which is
evidenced by the strong intensity of the [0001] pole, the β-phase heat treatment diminished the preferential distribution of poles significantly. This is evidenced by the uniform distribution of the pole intensities in the β-phase heat treated specimen as shown in Fig. 3 (c). The visual difference in the texture characteristics of the three specimens shown in the stereographic projections was confirmed by a quantitative estimate of the relative basal pole intensity along the sheet normal. Thus, the $f_n$ numbers were 0.643, 0.743 and 0.321 for the as-received condition, the 50 % cold-rolled plus α-phase heat treatment condition and the β-phase heat treatment condition, respectively.

Cold rolling varied the texture characteristics of Zircaloy-4 sheet in two ways. First, the amount of cold work increased the basal texture. The $f_n$ number increased as the sheet underwent 42.5 % rolling and increased further after 50 % rolling. Second, the mode of cold working also influenced the texture. Of the two rolling methods studied cross rolling caused a greater increase in the basal texture than direct rolling. The effect of cold rolling on the $f_n$ number is illustrated in Fig. 4 as a function of both the amount of cold work and the rolling method.

The effect of heat treating temperature also affected the texture characteristics of Zircaloy-4 sheet significantly. Above the α-β transus a reorientation of the unit cells accompanied the phase transformation. Below the transus α-phase annealing at a higher temperature resulted in a stronger basal texture along the sheet normal. This conclusion was obtained from heat treating experiments at 400° C, 550° C, and 1000° C, respectively, the results of which are shown in Fig. 5. As shown in Fig. 3 the β-phase heat treatment decreased the texture remarkably, randomizing the pole distribution. A cooling rate effect, albeit small, was also observed. For example, oil quenching randomized the texture more effectively than ice water quenching after β-phase heat treatment. The results of measurements of the $f_n$ numbers of variously treated specimens
are presented in Fig. 6, which demonstrates that rolling and heat treatments described above have a statistical significance despite data scatter.

The effects of cold rolling and heat treatments on the texture characteristics of Zircaloy-4 sheet as analyzed by the inverse pole figure and $f_n$ number were checked by the direct pole figure. Quarter-plane direct pole figures were obtained for [0001] poles with the sheet normal as a reference direction. The results of this study of the distribution characteristics of the basal poles were in general agreement with the results of the inverse pole figure in that the cold rolling and $\alpha$-phase heat treatment increased the intensity of the basal poles along the sheet normal direction while the $\beta$-phase heat treatment decreased it. Two representative direct basal pole figures and corresponding $F_n$ numbers of the as-received specimen and the $\beta$-phase-heat-treated specimen are shown in Fig. 7. The dispersion of the basal pole intensity in the $\beta$-phase-heat-treated specimen is evident in contrast to the preferential alignment of the basal poles along the sheet normal in the as-received condition.

Variation in the crystallographic texture in Zircaloy-4 sheet is expected to result in a variation in the mechanical anisotropy. The mechanical anisotropy was analyzed by the yield loci which were constructed from the microhardness numbers. The yield loci were obtained for each specimen and those of three selected specimens are shown in Fig. 8. From the yield loci it may be concluded that the mechanical anisotropy increased with cold rolling or $\alpha$-phase heat treatment while it decreased after $\beta$-phase heat treatment, which is in qualitative agreement with the result of the X-ray diffraction study. It is noted that a slight anisotropic softening occurred along the sheet normal direction in the $\beta$-phase heat treated specimen. The anisotropy parameter, $R$ value, was obtained by analyzing the yield loci with the aid of Eq. (1). A summary of $f_n$, $F_n$ and $R$ values of all the specimens studied are presented in Table I.
The anisotropy parameters, R, of cold rolled and heat treated Zircaloy-4 specimens are shown in Fig. 9 and Fig. 10 as a function of the amount of cold rolling and heat treating temperature, respectively. The trend in the variation of the mechanical anisotropy of Zircaloy-4 sheet due to processing condition was very similar to that of the texture coefficients shown in Fig. 3 and Fig. 4.

Figure 11 shows the relationship between the texture parameter $f_n$ and the anisotropy parameter $R$. As expected, the mechanical anisotropy parameter $R$ increased monotonically with $f_n$. A similar trend was observed between $R$ and $F_n$. Van Swam et al.\(^{(13)}\) suggested a functional relationship between the texture parameters and the anisotropy parameter of Zircaloy tubing, where the $R$ values were determined by uniaxial tensile testing along the tube axis. The experimental data points and the theoretical prediction curve by these authors are given in Fig. 11 along with the data points from the present work on Zircaloy-4 sheet. There is a fair agreement between the experimental values and the theoretical prediction of the two parameters especially for the severely textured materials. The experimental measurements of the $R$-values, however, tend higher than the theoretical prediction and this trend was more pronounced in less severely textured materials.

4. DISCUSSION

As shown in Fig. 3, the basal pole intensity along the sheet normal increased with the amount of cold work regardless of the rolling method. This phenomenon is consistent with the general behavior of hcp crystals with $c/a$ ratio less than the ideal value, in that, the $c$ axis of the unit cell tends to align with the direction of the compressive strain. The
normal basal texture in sheet is similar to the radial basal texture in tubing. A slight difference was observed, however, in that the maximum intensity of the basal poles in the rolled sheet tested here was along the normal direction whereas the bipolar maxima usually occur at 30 to 40° from the radial direction in tubing\(^\text{[19-20]}\). This may be due to an insertion of cross rolling in the manufacturing schedule, the details of which is not known.

The fact that the cross rolling method resulted in a stronger normal basal texture is probably due to the rotation of the \(c\) axis caused by twinning during plastic deformation. During the first direct rolling the unit cells whose \(c\) axes lie in the \(r-n\) plane tend to rotate toward the sheet normal. The rotation may occur early in the deformation process after as little as 1.5% plastic strain as Ballinger observed.\(^{\text{21}}\) Rolling is essentially a biaxial plastic deformation along the sheet normal and the rolling direction. Different rolling directions, therefore, define different crystallographic planes and directions whose Schmid factors are favorable for twinning. Since cross rolling involves two different rolling directions in contrast to a single rolling direction in direct rolling the former activates more twinning system than the latter. The unit cells whose \(c\) axes rotate so that the basal poles align with the sheet normal resist further rotation. As a consequence, the normal basal texture increases with the amount of cold work, and more basal poles rotate toward the sheet normal during cross rolling than direct rolling.

As shown by Fig. 11 the qualitative relationship between \(R\) and \(f_n\) or \(R\) and \(F_n\) obtained experimentally from the present work and others followed the trend predicted by the model of Van Swam et al.\(^{\text{13}}\) This is interesting because the shape of the experimental material and the technique to obtain \(R\) values were different in the two studies. It is noted however, that the \(R\) values definitely show a positive variance from
the model prediction which is most pronounced for low \( f_n \) values. The reason for the discrepancy may be over-simplification of the pole distribution.

The model of Van Swam et al. based on single crystal approximation and prism slip mechanism may be expressed as follows:

\[
\begin{align*}
\frac{f_n}{R + 1} & = R - 1 \quad \text{(2)} \\
\frac{F_n}{R + 1} & = \frac{R - 1}{R + 1} \quad \text{(3)}
\end{align*}
\]

In this model all the basal poles are assumed to lie on the single radial-tangential plane of tubing which is equivalent to the \( n-t \) plane of sheet in the present study. The experimental values of the texture numbers, \( f_n \) and \( F_n \), on the other hand, are obtained by the following equations:

\[
\begin{align*}
\text{Kearns number, } f_n & = \int_{0}^{+\pi/2} I(\phi) \sin\phi \cos^2\phi \, d\phi \\
\text{Kallstrom number, } F_n & = \frac{\int_{-\pi/2}^{+\pi/2} I[0001](\phi) \cos^2\phi \, d\phi}{\int_{-\pi/2}^{+\pi/2} I[0001](\phi) \, d\phi}
\end{align*}
\]

where \( \phi \) is the angle between the basal pole and the sheet normal. It is noted that the meaning of \( \phi \) in equations (4) and (5) is not restricted to the \( n-t \) plane but broadened into a three dimensional distribution. In the computation procedure in the experiments, therefore, the intensity of the basal poles having an angle with the \( n-t \) plane is included. This would result in an \( f_n \) value of 1/3 corresponding to the isotropic material with \( R = 1 \) whereas \( f_n = 1/2 \) is expected from Eq. (2). The normalizing process in the computation of the \( f_n \) number is expected to result in a more pronounced deviation of the \( R \) values for the low \( f_n \) regime since the number of the poles aligned to the sheet normal is relatively low.
5. CONCLUSIONS

The following conclusions were drawn from this study of the mechanical anisotropy and the crystallographic texture of Zircaloy-4 sheet:

(1) The basal texture along the sheet normal can be increased by either cold rolling, particularly cross rolling, or α-phase annealing heat treatment following cold rolling.

(2) The relationship between the crystallographic texture coefficient ($f_n$ or $F_n$) and the mechanical anisotropy parameter ($R$) followed the trend predicted by the model of Van Swam et al. qualitatively. Quantitatively, however, the experimental $R$ values were higher than the theoretical prediction, especially for lower $f_n$ values. The reason was attributed to the assumption of the basal pole distribution on a single plane in the model.

(3) The pole distribution was significantly randomized and Zircaloy sheet was slightly softened along the normal direction by β-quenching heat treatment at $1000^\circ$ C.

6. ACKNOWLEDGEMENT

The authors are indebted to Dr. M. Yang of the Korea Atomic Energy Research Institute for Zircaloy-4 test materials. We also wish to extend thanks to Drs. R.J. Comstock, G.P. Sabol, and K.L. Murty for the critical discussions on the subject and to Mr. Y. Im for the valuable help in manuscript preparation. This work was performed under the auspices of the Korean Science and Engineering Foundation, and partly supported by the Director, Office of Energy Research, Office of Basic Energy Science, Materials Sciences Division of the U.S. Department of Energy under contract # DE-AC03-76SF00098.
7. REFERENCES

Table I. Texture Coefficients and Anisotropy Parameters of Zircaloy-4 sheet

<table>
<thead>
<tr>
<th>Treatment</th>
<th>f_n</th>
<th>F_n</th>
<th>R</th>
</tr>
</thead>
<tbody>
<tr>
<td>As-received</td>
<td>0.643</td>
<td>0.237</td>
<td>1.77</td>
</tr>
<tr>
<td>Direct Rolling, 42.5%</td>
<td>0.662</td>
<td>0.275</td>
<td>1.93</td>
</tr>
<tr>
<td>Direct Rolling, 50%</td>
<td>0.689</td>
<td>0.319</td>
<td>2.14</td>
</tr>
<tr>
<td>Cross Rolling, 42.5%</td>
<td>0.669</td>
<td>0.292</td>
<td>2.00</td>
</tr>
<tr>
<td>Cross Rolling, 50%</td>
<td>0.703</td>
<td>0.301</td>
<td>2.38</td>
</tr>
<tr>
<td>Direct Rolling, 50%+400°C/1.5h/FC</td>
<td>0.725</td>
<td>0.378</td>
<td>2.72</td>
</tr>
<tr>
<td>Direct Rolling, 50%+550°C/1.5h/FC</td>
<td>0.743</td>
<td>0.449</td>
<td>2.83</td>
</tr>
<tr>
<td>Direct Rolling, 50%+950°C/1.5h/1Q</td>
<td>0.692</td>
<td>0.316</td>
<td>2.21</td>
</tr>
<tr>
<td>Cross-Rolling, 50%+400°C/1.5h/FC</td>
<td>0.745</td>
<td>0.533</td>
<td>3.28</td>
</tr>
<tr>
<td>Cross Rolling, 50%+550°C/1.5h/FC</td>
<td>0.751</td>
<td>0.546</td>
<td>3.49</td>
</tr>
<tr>
<td>Cross Rolling, 50%+950°C/1.5h/1Q</td>
<td>0.685</td>
<td>0.315</td>
<td>2.04</td>
</tr>
<tr>
<td>As-received+1000°C/1h/OQ</td>
<td>0.321</td>
<td>-0.189</td>
<td>0.63</td>
</tr>
<tr>
<td>As-received+1000°C/1h/1Q</td>
<td>0.381</td>
<td>-0.163</td>
<td>0.82</td>
</tr>
</tbody>
</table>
FIGURE CAPTIONS

Fig.1 Reference directions of rolled Zircaloy-4 sheet

Fig.2 (0001) stereographic projection of the hcp unit cell used for the inverse pole figure

Fig.3 Inverse pole figures of Zircaloy-4 sheet in (a) as-received condition, (b) cross rolled 50% and annealed (550°C/1.5 h/FC) condition, and (c) as-received and annealed (1000°C/1.5 h/FC) condition

Fig.4 Effect of cold rolling on the basal texture of Zircaloy-4 sheet along the sheet normal direction: cr: cross rolling, dr: direct rolling

Fig.5 Kearns number ($f_n$) of cold worked Zircaloy-4 sheet as a function of annealing temperature: cr: cross rolled, dr: direct rolled

Fig.6 Kearns numbers of variously processed Zircaloy-4 sheet: AR: as-received, DR + 550: direct rolled 50% and annealed 550°C/1.5 h, CR + 550: cross rolled 50% and annealed 550°C/1.5 h

Fig.7 (0001) direct pole figures of (a) as-received and (b) beta-quenched (as-received + 1000°C/1 h/OQ) Zircaloy-4 sheet

Fig.8 Yield loci of cold-rolled and heat-treated Zircaloy-4 sheet with the units indicating Knoop hardness number

Fig.9 Anisotropy parameter $R$ of Zircaloy-4 sheet as a function of the amount of cold work

Fig.10 Anisotropy parameter $R$ of Zircaloy-4 sheet as a function of annealing temperature

Fig.11 Anisotropy parameter $R$ of Zircaloy-4 sheet as a function of the Kearns number $f_n$
Fig. 1 The reference directions of rolled Zircaloy-4 sheet
Fig. 2 (0001) stereographic projection of the hcp unit cell used for the inverse pole figure
Fig. 3  Inverse pole figures of Zircaloy-4 sheet in (a) as-received condition, (b) cross-rolled 50% and annealed (550° C/1.5 h/FC) condition, and (c) as-received and annealed (1000° C/1.5 h/FC) condition.
Fig. 4 Effect of cold rolling on the basal texture of Zircaloy-4 sheet along the sheet normal direction: cr: cross rolling, dr: direct rolling
Fig. 5  Kearns number ($f_n$) of cold worked Zircaloy-4 sheet as a function of annealing temperature: cr: cross rolled, dr: direct rolled
Fig. 6 Kearns numbers of variously processed Zircaloy-4 sheet: AR: as-received, DR + 550: direct rolled 50% and annealed 550°C/1.5 h, CR + 550: cross rolled 50% and annealed 550°C/1.5 h
Fig. 7 (0001) direct pole figures of (a) as-received and (b) beta-quenched (as-received + 1000 °C/1 h/OQ) Zircaloy-4 sheet.
Fig. 8 Yield loci of cold-rolled and heat-treated Zircaloy-4 sheet with the units indicating Knoop hardness number
Fig. 9 Anisotropy parameter $R$ of Zircaloy-4 sheet as a function of the amount of cold work.
Fig. 10 Anisotropy parameter $R$ of Zircaloy-4 sheet as a function of annealing temperature
Fig. 11 Anisotropy parameter $R$ of Zircaloy-4 as a function of the Kearns number $f_n$

$$R = \frac{f_n}{(1 - f_n)}$$